

B. Amendment to the Specification

Please amend the paragraph on page 1, lines 5-12, as follows:

--The present invention relates to a luminescence device and a metal coordination compound therefor. More specifically, the present invention relates to a luminescence device employing an organic metal coordination compound having a platinum center metal as a luminescence material so as to allow stable luminescence efficiency, and a metal coordination compound adapted for use in the luminescence device.--

Please amend the paragraph on page 2, lines 14-22, as follows:

--The luminescence layer 12 may be formed of, e.g., an aluminum quinolinol complex (representative example thereof may include Alq3 described hereinafter) having an electron transporting characteristic and a luminescent characteristic. The hole transport layer 13 may be formed of, e.g., a triphenyldiamine derivative (representative example thereof may include α -NPD described hereinafter) having an electron donating characteristic.--

Please amend the paragraph on page 3, lines 11-17, as follows:

--Referring to Figure 1B, in addition to the layers shown in Figure 1A, an electron transport layer 16 is disposed between the metal electrode 11 and the luminescence layer 12, whereby an effective carrier blocking performance can be ensured

by separating the functions of luminescence, electron transport and hole transport, thus allowing effective luminescence.--

Please amend the paragraphs on page 3, line 20 - page 4, line 6, as follows:

--In ordinary organic EL devices, fluorescence caused during a transition of a luminescent center molecule from a singlet excited state to a ground state is used as luminescence.

On the other hand, not the above fluorescence (luminescence) via singlet exciton, but phosphorescence (luminescence) via a triplet exciton has been studied for use in an organic EL device as described in, e.g., "Improved energy transfer in electrophosphorescent device" (D.F. O'Brien et al., Applied Physics Letters, Vol. 74, No. 3, pp. 442 - 444 (1999)) and "Very high-efficiency green organic light-emitting devices based on electrophosphorescence" (M.A. Baldo et al., Applied Physics Letters, Vol. 75, No. 1, pp. 4 - 6 (1999)).--

Please amend the paragraphs on page 6, line 19 - page 7, line 11, as follows:

--More specifically, in the case of the phosphorescence material, excitons produced by a recombination of carriers comprise singlet excitons and triplet excitons presented in a ratio of 1:3. For this reason, when fluorescence caused during the transition from the singlet excited state to the ground state is utilized, a resultant luminescence efficiency is 25 % (as upper limit) based on all the produced excitons in principle.

On the other hand, in the case of utilizing phosphorescence caused during a transition from the triplet excited state, a resultant luminescence efficiency is expected to be at least three times that of the case of fluorescence in principle. In addition thereto, if an intersystem crossing from the singlet excited state (higher energy level) to the triplet excited state is taken into consideration, the luminescence efficiency of phosphorescence can be expected to be 100 % (four times that of fluorescence) in principle.--

Please amend the paragraphs on page 7, line 17 - page 8, line 7, as follows:

--However, the above-mentioned organic EL devices utilizing phosphorescence have ~~accompanied with~~ a problem associated with of luminescent deterioration particularly in an energized state.

The reason for luminescent deterioration has not been clarified as yet but may be attributable to such a phenomenon that the life of triplet exciton is generally longer than that of singlet exciton by at least three digits, so that a molecule is placed in a higher-energy state for a long period, causing a ~~to cause~~ reaction with an ambient substance, formation of exciplex or excimer, a change in a minute molecular structure, a structural change of an ambient substance, etc.

Accordingly, the (electro)phosphorescence EL device is expected to provide a higher luminescence efficiency as described above, while the EL device is required to suppress or minimize the luminescent deterioration in the energized state.--

Please amend the paragraph on page 8, lines 10-14, as follows:

--An object of the present invention is to provide a luminescence device capable of providing a high-efficiency luminescent state at a high brightness (or luminance) for a long period while minimizing the deterioration in luminescence in the energized state.--

Please amend the paragraph on page 9, line 22 - page 10, line 11, as follows:

--In the case where a luminescence layer for an organic EL device is formed of a carrier transporting host material and a phosphorescent guest material, a process of emission of light (phosphorescence) may generally involve the following steps:

- (1) transport of an electron and a hole within a luminescence layer,
- (2) formation of an exciton of the host material,
- (3) transmission of excited energy between host material molecules,
- (4) transmission of excited energy from the host material molecule to the guest material molecule,
- (5) formation of a triplet exciton of the guest material, and
- (6) emission of light (phosphorescence) caused during a transition from the triplet excited state to the ground state of the guest material.--

Please amend the paragraphs on page 10, line 15 - page 11, line 12, as follows:

--In order to improve a luminescence efficiency of the EL device, a luminescence center material per se is required to provide a higher yield of a luminescence

quantum. In addition thereto, an efficient energy transfer between host material molecules and/or between host material molecule and guest material molecule is also an important factor.

Further, the above-described luminescent deterioration in an energized state may presumably relate to the luminescent center material per se or an environmental change thereof by its ambient molecular structure.

For this reason, our research group has extensively investigated an effect of the use of the metal coordination compound (platinum complex) having a partial structure of formula (1) as the luminescent center ~~luminescentcenter~~ material, and as a result[[,]] has found that the metal coordination compound having the partial structure of formula (1) allows a high-efficiency luminescence (e.g., luminescence efficiency of at least 1 cd/W) with a high brightness (luminance) for a long period (e.g., a luminance half-life of at least 500 hours at an initial luminance of 100 cd/m²) (i.e., a decreased luminescent deterioration in an energized state).--

Please amend the paragraph on page 12, lines 23-27, as follows:

--The metal coordination compound may more preferably be represented by the formula (1-1) or the formula (1-2) in order to allow further improved high-efficient luminance while minimizing the luminescent deterioration in the energized state.--

Please amend the paragraph on page 13, lines 11-19, as follows:

--The metal coordination compound (platinum complex) specifically represented by the above formulas (1-1) to (1-6) causes phosphorescence (luminescence) and is assumed to have a lowest excited state comprising a triplet excited state likely liable to cause a metal-to-ligand charge transfer (MLCT* state). The phosphorescent emission of light (phosphorescence) is produced during the transition from the MLCT* state to the ground state.--

Please amend the paragraphs on page 13, line 24 - page 14, line 18, as follows:

--A phosphorescence yield (P(m)) is obtained based on the following equation:

$$P(m)/P(s) = (S(m)/S(s)) \times (A(s)/A(m)),$$

wherein P(m) represents a phosphorescence yield of an (unknown) objective luminescent material, P(s) represents a known (standard) phosphorescence yield of a standard luminescent material (Ir(ppy)₃), S(m) represents an integrated intensity of (photo-)excited emission spectrum of the objective material, S(s) represents a known integrated intensity of the standard material, A(m) represents an absorption spectrum of an excited light wavelength of the objective material, and A(s) represents a known absorption spectrum of the standard material.

The shorter phosphorescence life is necessary to provide a resultant EL device with a higher luminescence efficiency. This is because the longer phosphorescence life increases molecules placed in their triplet excited state, which is a waiting state for

phosphorescence, thus lowering the resultant luminescence efficiency particularly at a higher current density.--

Please amend the paragraphs on page 14, line 24 - page 17, line 1, as follows:

--In a conventional phosphorescent EL device, ~~uses~~ the platinum-porphiline complex (e.g., PtOEP described above) is used as the luminescent material. On the other hand, the metal coordination compound according to the present invention has a carbon-platinum bond (C-Pt bond) in its molecular structure, thus particularly effectively exhibiting a heavy atom effect of platinum (Pt) compared to the case of N-Pt bond (in PtOEP). As a result, a spin-orbit interaction is enhanced to realize a higher phosphorescence yield and a shorter phosphorescence life at the same time.

Further, molecules of the metal coordination compound have a shorter time period wherein they stay in the triplet excited state, thus prolonging the life of the EL device with less deterioration. In this regard, the metal coordination compound according to the present invention has been substantiated to exhibit excellent stability of luminance as shown in the Examples ~~described~~ hereinafter.

In the case of a phosphorescent (luminescent) material, luminescent characteristics are largely affected by the ~~its~~ molecular environment. On the other hand, principal characteristics of the fluorescent material are studied based on photoluminescence.

For this reason, results of photoluminescence of the phosphorescent material do not reflect the luminescent characteristics of the resultant EL device in many cases since the luminescent characteristics in the case of the phosphorescent material depend on a magnitude of the polarity of the ambient host material molecules, the ambient temperature, and the presence state of the material (e.g., solid state or liquid state, etc.) Accordingly, different from the fluorescent material, it is generally difficult to expect the resultant EL characteristics for the phosphorescent material by simply removing a part of the characteristics from photoluminescence results.

As a feature of the molecular structure, the platinum complex has a planar structure, and the energy transfer of triplet exciton (i.e., energy transfer from the host material molecule in the triplet excited state to the guest material molecule) is performed based on the electron exchange between adjacent molecules (so-called Dexter transfer). Accordingly, a degree of overlapping of the electron clouds ~~cloud~~ between adjacent molecules is an important factor, so that the planar (molecular) structure is suitable for an efficient energy transfer.

On the other hand, Ir(ppy)₃ (iridium-phenylpyrimidine complex) as used in the above-described conventional EL device has a steric octahedral coordination structure, thus failing to perform an efficient energy transfer (Dexter transfer) from the host material molecule.

As described above, the metal coordination compound (platinum complex) according to the present invention is a suitable luminescent material for an EL device.--

Please amend the paragraph on page 17, line 21 - page 18, line 18, as follows:

--The luminescence device according to the present invention may be applicable to devices required to allow energy saving and high luminance, such as those for a display apparatus and an illumination apparatus, a light source for printers, and a backlight (unit) for a liquid crystal display apparatus. Specifically, in the case of using the luminescence device of the present invention in the display apparatus, it is possible to provide a flat panel display apparatus capable of exhibiting an excellent energy saving performance, a high visibility and a good lightweight property. With respect to the light source, it becomes possible to replace a laser light source of a laser beam printer, which is currently used widely, with the luminescence device according to the present invention. Further, when the luminescence device of the present invention is arranged in independently addressable arrays as an exposure means for effecting desired exposure of light to a photosensitive drum for forming an image, it becomes possible to considerably reduce ~~reducing~~ the volume (size) of the image forming apparatus. With respect to the illumination apparatus and the backlight (unit), the resultant apparatus (unit) using the luminescence device of the present invention is expected to have an energy saving effect.--

Please amend the paragraph on page 19, lines 19-24, as follows:

--In the above structural formulas, an unconnected covalent (single) linkage extended from nitrogen atom (N) in a lower-right direction except for Pz' is a linkage

connected to the platinum atom (Pt), and the other unconnected covalent linkage is a linkage connected to an adjacent cyclic group.--

Please amend the paragraph on page 21, lines 21-27, as follows:

--In the above structural formulas (Ph to Pz and Ph' to Pz'), an unconnected covalent (single) linkage extended in an upper-right direction is a linkage connected to the platinum atom (Pt), and the other unconnected covalent linkage extended in an upper direction is a linkage connected to an adjacent cyclic group.--

Please amend the paragraph on page 29, lines 1-4, as follows:

--Of the metal coordination compound preferably having the above-mentioned formulas (1-1) to (1-6), those of formulas (1-1) and (1-2) may, e.g., be synthesized through the following reaction ~~reacton~~ schemes.--

Please amend the paragraph on page 32, lines 7-9, as follows:

--Each of the luminescence devices having a structure shown in Figure 1B was ~~were~~ prepared in the following manner.--

Please amend the paragraphs on page 33, lines 3-17, as follows:

--Each of the thus-prepared luminescence devices was taken out of the vacuum chamber and was subjected to a continuous energization test in an atmosphere of a

dry nitrogen gas stream so as to remove device deterioration factors, such as oxygen and moisture (water content).

The continuous energization test was performed by continuously applying a voltage at a constant current density of 70 mA/cm² to the luminescence device having the ITO (transparent) electrode (as an anode) and the Al (metal) electrode (as a cathode), followed by a measurement of luminance (brightness) with time so as to determine a time (luminance half-life) required for decreasing an initial luminance (80 - 120 cd/m²) to ½ thereof.--

Please amend the paragraph on page 35, line 14 - page 36, line 10, as follows:

--In a 3 liter-three necked flask, 14.6 g (90.6 mM) of 2-(2-thienyl)pyridine and 912 ml of anhydrous ether were placed and stirred at -70°C or below in an argon gas stream. To the mixture, 62.2 ml (99.5 mM) of 1.6M-t-butyllithium solution in pentane was added dropwise in ca. 35 min., followed by stirring at -70°C for 40 min. At that temperature, a suspension of 8.5 g (19.0 mM) of cis-PtCl₂[(C₂H₅)₂S]₂ in a mixture solvent of 289 ml of anhydrous ether and 73 ml of tetrahydrofuran (THF) was added dropwise in ca. 1 hour to the resultant mixture, followed by stirring at -70°C for 30 min. and a gradual temperature rise up to 0°C in ca. 2 hours. To the reaction mixture, 912 ml of water was gradually added dropwise at 0°C. The organic layer was washed with common salt aqueous solution and the aqueous (water) layer was subjected to extraction with methylene chloride. The resultant organic layer (from the organic and aqueous layers) was dried with

anhydrous sodium sulfate, followed by distilling off ~~distilling off~~ of the solvent to obtain a residue. The residue was recrystallized from a mixture solvent (hexane/methylene chloride) to obtain 4.50 g of cis-bis[2-(2-thienyl)pyridinato-N,C^{5'}] platinum (II) (Yield: 45.8 %).--

Please amend the paragraphs on page 36, line 16 - page 37, line 10, as follows:

--In a 1 liter-three necked flask, 26.6 g (168.5 mM) of 2-bromopyridine, 30.0 g (168.5 mM) of benzo[b]thiophene-2-boronic acid, 170 ml of toluene, 85 ml of ethanol and 170 ml of 2M-sodium carbonate aqueous solution were placed and stirred in a nitrogen gas stream at room temperature. Under stirring, to the mixture, 6.18 g (5.35 mM) of tetrakis(triphenyl-phosphine) palladium (0) was added, followed by heat-refluxing for 5.5 hours under stirring in a nitrogen gas stream.

After the reaction, the reaction mixture was cooled, followed by extraction with cool water and toluene. The organic layer was washed with water until the system showed neutral, followed by distilling off of the solvent under reduced pressure to obtain a residue. The residue was purified by silica gel column chromatography (eluent: toluene/hexane = 5/1) to obtain a colorless crystal. The crystal was purified by alumina column chromatography (eluent: toluene) and recrystallized from ethanol to obtain 12.6 g of 2-(pyridine-2-yl)benzo[b]thiophene (Yield: 35.4 %).--

Please amend the paragraph on page 37, line 17 - page 38, line 13, as

follows:

--In a 3 liter-three necked flask, 6.73 g (31.9 mM) of 2-(benzo[b]thiophene-2-yl)pyridine and 636 ml of anhydrous ether were placed and stirred at -70°C or below in an argon gas stream. To the mixture, 21.9 ml (35.0 mM) of 1.6M-t-butyllithium solution in pentane was added dropwise in ca. 20 min., followed by stirring at -70°C for 50 min. At that temperature, a suspension of 2.97 g (6.68 mM) of cis-PtCl₂[(C₂H₅)₂S]₂ in a mixture solvent of 101 ml of anhydrous ether and 25 ml of tetrahydrofuran (THF) was added dropwise in ca. 30 min. to the resultant mixture, followed by stirring at -70°C for 1 hour[[.]] and a gradual temperature rise up to 0°C in ca. 2 hours. To the reaction mixture, 318 ml of water was gradually added dropwise at 0°C . The organic layer was washed with a common salt aqueous solution and the aqueous (water) layer was subjected to extraction with methylene chloride. The resultant organic layer (from the organic and aqueous layers) was dried with anhydrous sodium sulfate, followed by distilling off ~~distilling-off~~ of the solvent to obtain a residue. The residue was recrystallized from a mixture solvent (hexane/methylene chloride) to obtain 3.10 g of cis-bis[2-(benzo[b]thiophene-2-yl)pyridinato-N,C^{5'}] platinum (II) (Yield: 75.4 %).--

Please amend the paragraph on page 38, line 21 - page 39, line 17, as

follows:

--In a 3 liter-three necked flask, 35.0 g (112 mM) of 2,2'-dibromobiphenyl and 650 ml of anhydrous ether were placed and stirred at -60°C or below in an argon gas

stream. To the mixture, 153 ml (0.245 mM) of 1.6M-n-butyllithium solution in pentane was added dropwise in ca. 50 min., followed by a temperature rise and stirring at room temperature for 3 hours. To a suspension of 25.0 g (56.0 mM) of cis-PtCl₂[(C₂H₅)₂S]₂ in 833 ml of anhydrous ether cooled and kept at -10° C or below, the resultant mixture was added dropwise in ca. 10 min., followed by stirring at -10°C for 1 hour and a gradual temperature rise up to 0°C. To the reaction mixture, 417 ml of water was gradually added dropwise at 0°C. The organic layer was washed with a common salt aqueous solution and the aqueous (water) layer was subjected to extraction with methylene chloride. The resultant organic layer (from the organic and aqueous layers) was dried with anhydrous sodium sulfate, followed by ~~distilling off~~ distilling-off of the solvent to obtain a residue. The residue was successively recrystallized from a mixture solvent (hexane/ether) and a mixture solvent (hexane/methylene chloride) to obtain 1.77 g of a compound (A) (Yield: 7.2 %).--

Please amend the paragraph on page 39, line 25 - page 40, line 9, as follows:

--In a 100 ml-three-necked flask, 21.3 g (136 mM) of 2,2'-dipyridyl was placed and melted at 80°C in an argon gas stream, followed by the addition of 1.73 g (1.98 mM) of the above-prepared compound (A). The mixture was stirred at 80°C for 10 min. under reduced pressure and cooled to ca. 10°C to crystallize the mixture. The crystallized mixture was dissolved in methylene chloride, and ~~thereto~~, hexane was added thereto to reprecipitate a crystal. The crystal was recovered by filtration, followed by recrystallization

from a mixture solvent (hexane/methylene chloride) to obtain 1.90 g of an objective compound (B) (Yield: 95.4 %).--

Please amend the paragraph on page 40, line 24 - page 41, line 2, as follows:

--The luminescence device (EL device) using the metal coordination compound according to the present invention allows a high-efficiency luminescence at a high luminescence for a long period of time while minimizing luminescence deterioration in an energized state.--